Workshop Summary & Conclusions

Cementitious Material for Waste Treatment, Disposal, Remediation and Decommissioning

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Consortium for Risk Evaluation with Stakeholder Participation

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A Team Effort!!

SRNL

Consortium for
Risk Evaluation with
Stakeholder Participation

VANDERBILT UNIVERSITY

EPA

WSRC

INERIS

cea

SCDHEC

SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

SRS CAB

NIST

Tuskegee University

WA Ecology

NM STATE UNIVERSITY

SRS

Laval University

ECN 50 YEARS
Energy research Centre of the Netherlands

Office of River Protection

CH2MHILL
Hanford Group, Inc.

UNIVERSITY OF ABERDEEN

University of Wisconsin-Madison

MINES PARIS

Battelle

CEREGE

Pacific Northwest National Laboratory
Operated by Battelle for the U.S. Department of Energy

Objectives and Expected Outcomes

Provide common understanding amongst DOE, regulators, site operators, researchers, and other stakeholders

- State of science
- Current practice
- Knowledge gaps

Identify opportunities to

- Improve waste management and restoration systems
- Reduce uncertainties in long-term performance conceptual models and quantitative predictions

C. Langton, D. Kosson

Motivation

Support DOE closure projects across the complex – waste treatment, containment structures, D&D, environmental restoration

- Grouting of tanks that contain residual radioactive materials
- Construction of vaults for waste containment/disposal
- Sealing and filling of voids in systems to be decommissioned
- Cementitious waste forms

Need for better estimates of long-term performance

- Near-surface disposal of radionuclides in cementitious materials and non-vitrified waste forms.
- Structural integrity & Isolation
- Release of constituents of concern

Citizens' Expectations

SRS Citizens Advisory Board Perspectives

- Regulatory-accepted monitoring with appropriate oversight.
- Peer-reviewed technology demonstrated at an adequate scale.
- Peer-reviewed performance prediction model.
- Contingency plan if performance does not meet requirements or predictions.

J. Ortaldo, SRS CAB

Example Project Needs
Relative Priority of Grout Attributes

Relative Priority	Attribute
Very high	**Low hydraulic conductivity**
Very high	**Degradation resistance**
High	**Pore water chemistry (↑pH, ↓Eh)**
Medium	**Strength**
Medium	**Flowability**

S. Reboul, J. Newman, SRS

Disposal System Functions Include:

Limit water contact with waste

Limit intruder contact with waste

Chemically retain radionuclides

Provide shielding

Stabilize waste (e.g., limit voids and erosion)

D. Esh, NRC

Role of Release Models in Performance Assessments

Water Infiltration

Residual Waste Testing

Contaminant Release Mechanism/Rate

Source Release Model

Source Release

Contaminant Fate & Transport Model

Hydrology & Sediment Geochemistry

Long-Term Performance Assessment

W. Deutsch, PNNL

EPA Response to Science Advisory Board Concerns

The Agency has also been seeking new testing approaches with:

- Better accuracy over a range of conditions
- Better foundation in basic science (i.e., not empirical)
- Better applicability in environmental assessments (i.e., groundwater fate and transport modeling)
- Flexibility to apply to a broad range of waste types and over a range of conditions that affect leaching and occur in management

Validation in both the lab and field

Practical applicability of tests

S. Thorneloe, G. Helms, USEPA

Overarching Leaching Assessment Framework

(Kosson, van der Sloot, Sanchez & Garrabrants, Environ. Engr. Sci., 19, 159-203, 2002)

Measure intrinsic leaching characteristics of material (aqueous-solid partitioning (pH and LS); release kinetics)

- Batch extractions & tank leaching (monoliths)
- Constituent fraction readily leached
- Controlling mechanism for release (mineral dissolution and solubility, solid phase adsorption, aqueous phase complexation)
- Release kinetics for mass transfer parameters

Evaluate release in the context of field scenario

- External influencing factors such as carbonation, oxidation
- Hydrology
- Mineralogical changes

Use geochemical speciation and mass transfer models to estimate release for alternative scenarios

- Model complexity to match information needs
- Many scenarios can be evaluated from single data set

Issues in Cementitious Materials Applications for Waste Isolation

- Issues are different depending on the duration (short vs. long half lives), magnitude, and characteristics of the hazard being mitigated.

- For commercial LLW disposal, most radionuclides were expected to decay to insignificant levels by 500 years

AND

- Justification of performance of cementitious materials beyond this was thought to be very challenging

D. Esh, NRC

Issues in Cementitious Materials Applications for Waste Isolation

- The time frame for regulatory analysis for waste isolation may extend to many thousands of years or beyond.
- The long time frame creates additional uncertainty, which may or may not be able to be addressed with research.

D. Esh, NRC

Example Uncertainties in Cementitious Materials Applications for Waste Isolation

- The hydrologic properties of cementitious materials over long time periods (> 100 years).
- Unsaturated properties of cementitious materials.
- The limited experience/database of retention properties of cementitious materials for some radionuclides (e.g., Sn-126, Se-79, Np-237).
- The degradation mechanisms and long-term performance of novel formulations (e.g., chemically engineered cements).

D. Esh, NRC

Example Uncertainties in Cementitious Materials Applications for Waste Isolation

- The validity of and assumed lack of synergism between the degradation mechanisms evaluated with the commonly used empirical relationships.
- The influence of fractures on degradation mechanisms.
- Oxidation of reducing formulations over time.
- Extension of laboratory-scale, short-term tests to large-scale, long-term applications (Does ANS 16.1 address mechanisms relevant to timeframes of 1000's of years?

D. Esh, NRC

A Few Research Suggestions (examples)

Areas of research that may be tractable are:

- Development of accelerated laboratory-scale test methods.
- Compilation of a database of international experience (both good and bad).
- Experiments to estimate the retention properties of cementitious materials for lesser studied radionuclides.
- Experiments to evaluate potential synergisms between degradation mechanisms, including the impact of fractures.

D. Esh, NRC

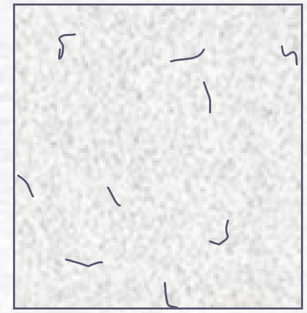
Many Coupled Processes: Integrated Long-Term Evolution

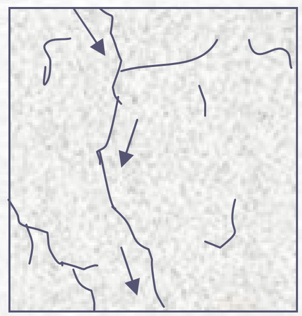
Chemical degradation and physical stress effects are coupled and integrated.

Physical stress
- Cyclic loading
- Flexural bending
- Drying shrinkage
- Seismic events
- Settlement

Chemical degradation
- Oxidation
- Leaching
- Expansive reactions
  - Carbonation
  - Sulfate attack
  - Rebar corrosion

Microcracks
- Increase porosity
- Increase interaction pore water/surface

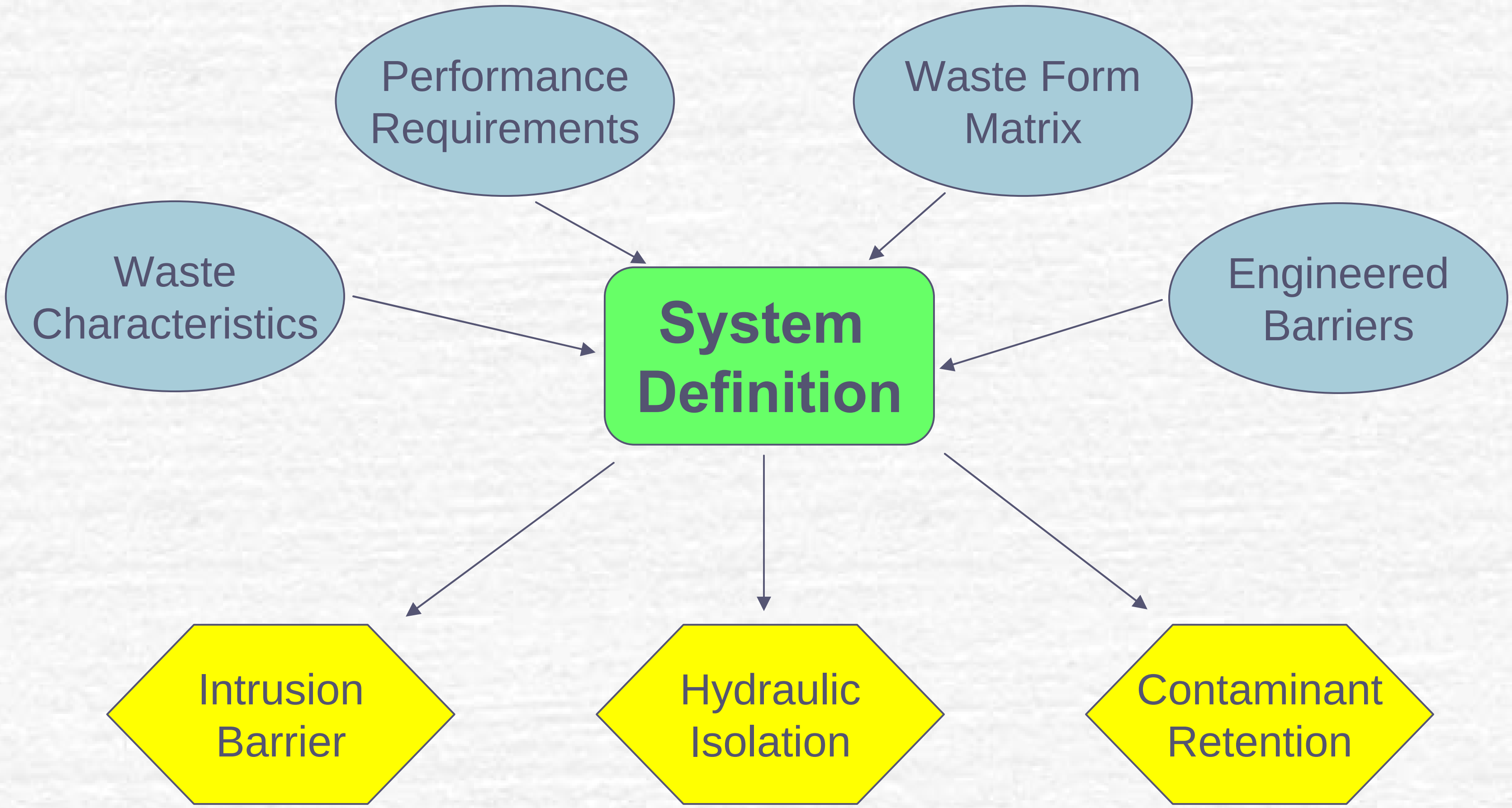




Through-cracks
- Preferential flow path
- Diffusive and convective release
- Loss of strength

Spalling
- Loss of cohesiveness
- Two body problem
- Eventual release from “granular” material

System Definition and Expectations



Understanding the System

System Definition

Boundary Conditions
Water, Oxygen
Carbon Dioxide
Sulfate, Chloride
Physical Stresses
Other wastes & external conditions

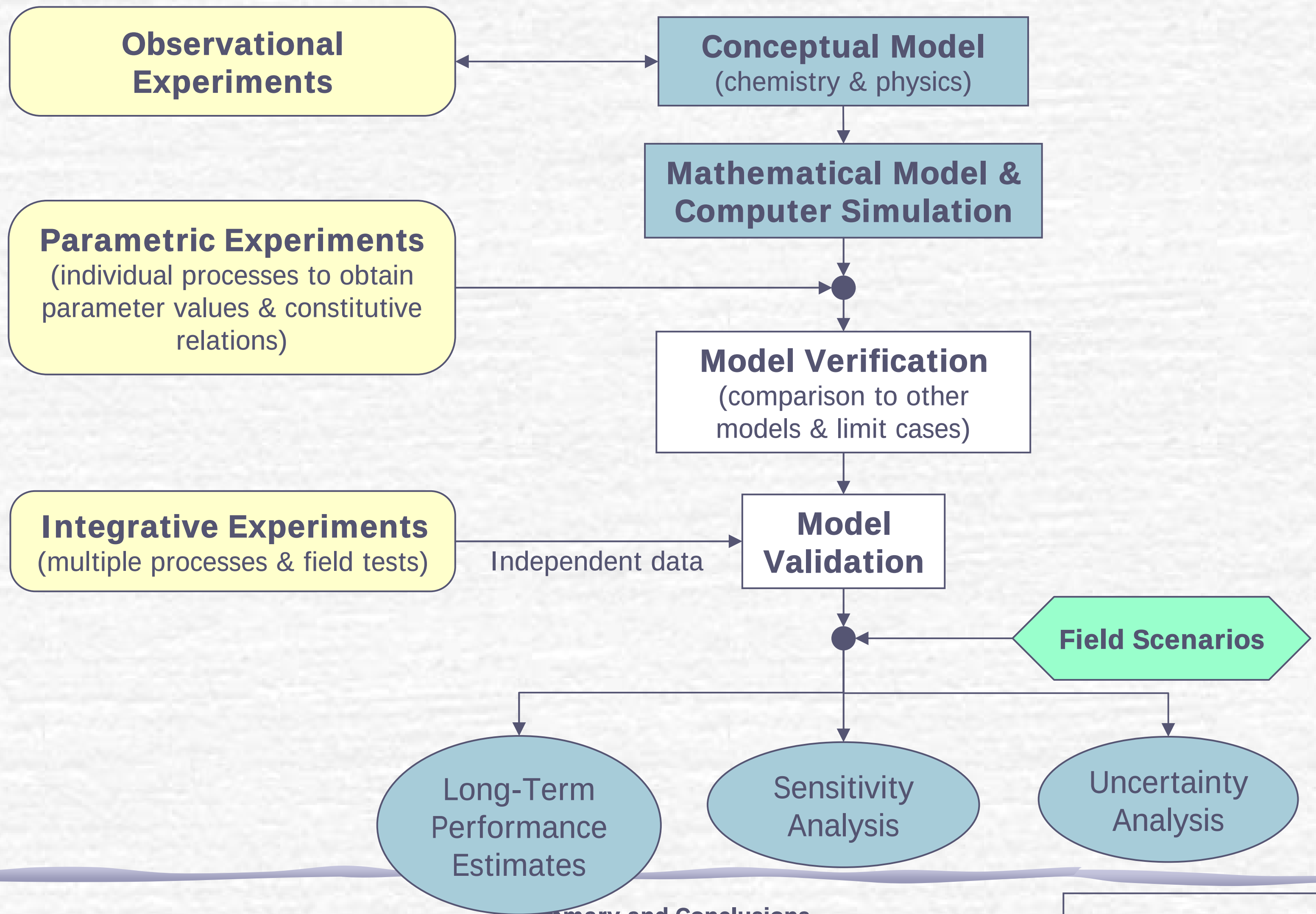
Define Controlling Degradation Mechanisms

Matrix Evolution
Permeability
Geochemistry
Aqueous & Gas Exchange
(advection, diffusion, capillary flow, barametric pumping)

Conceptual Model

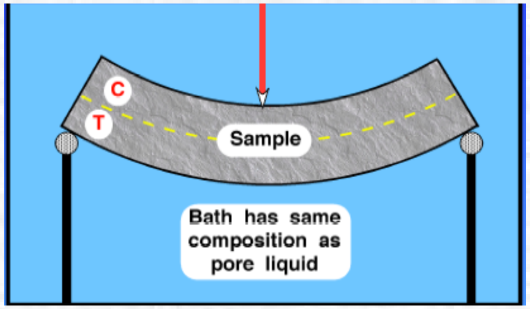
Source Release Model

Process- and Mechanism-Based Experimentation & Modeling

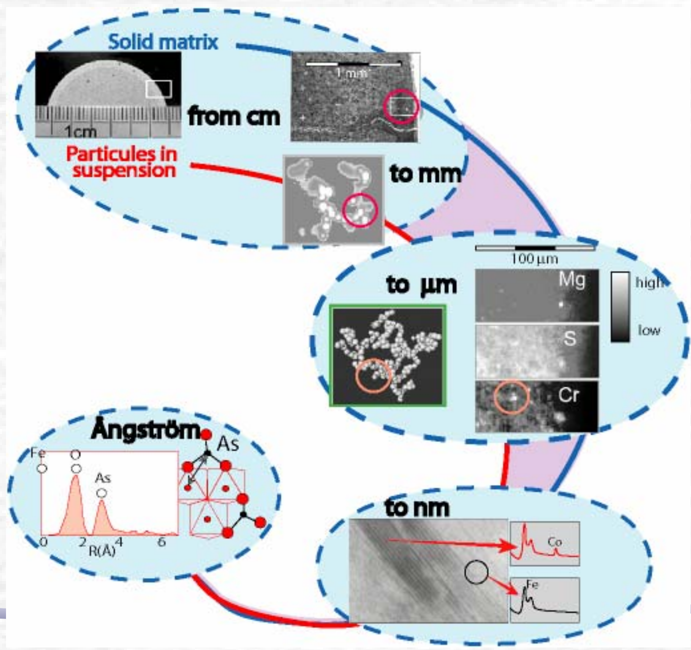


F. Sanchez, VU

Test Methods for Understanding & Implementation



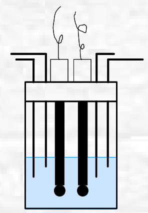
G. Shearer, Princeton



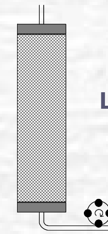
J. Rose, CEREGE

GRANULAR MATERIALS

or



pH DEPENDENCE TEST : BATCH MODE ANC TS 14429 or COMPUTER CONTROLLED TS 14997



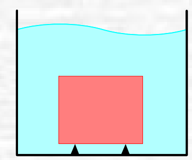
PERCOLATION LEACHING TEST (TS 14405)

MONOLITHIC MATERIALS

Crushed

Same as granular +

Intact



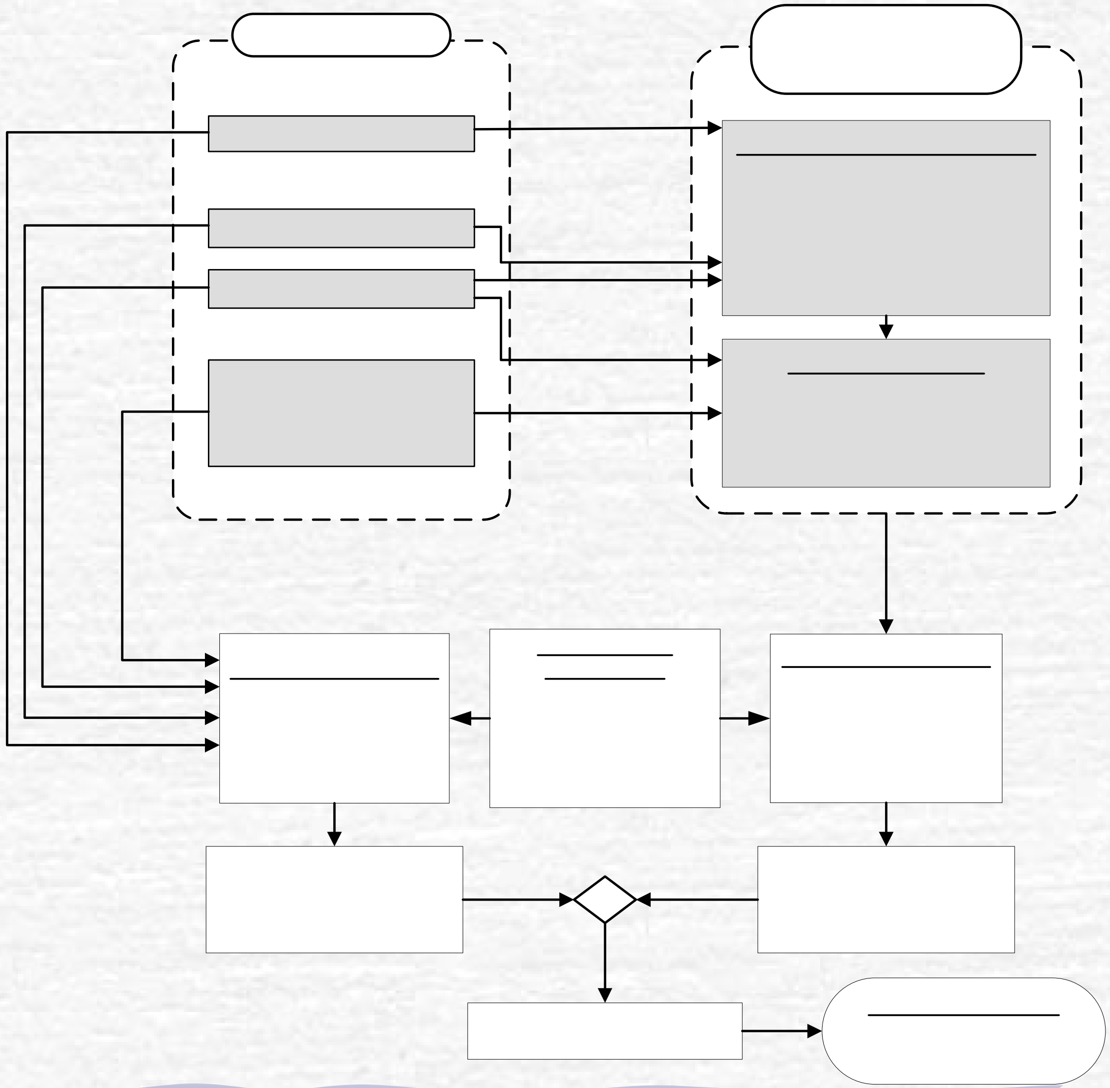
TANK LEACH TEST (MONOLITH) and COMPACTED GRANULAR LEACH TEST.

Chemical speciation aspects

Time dependent release

H. van der Sloot, ECN

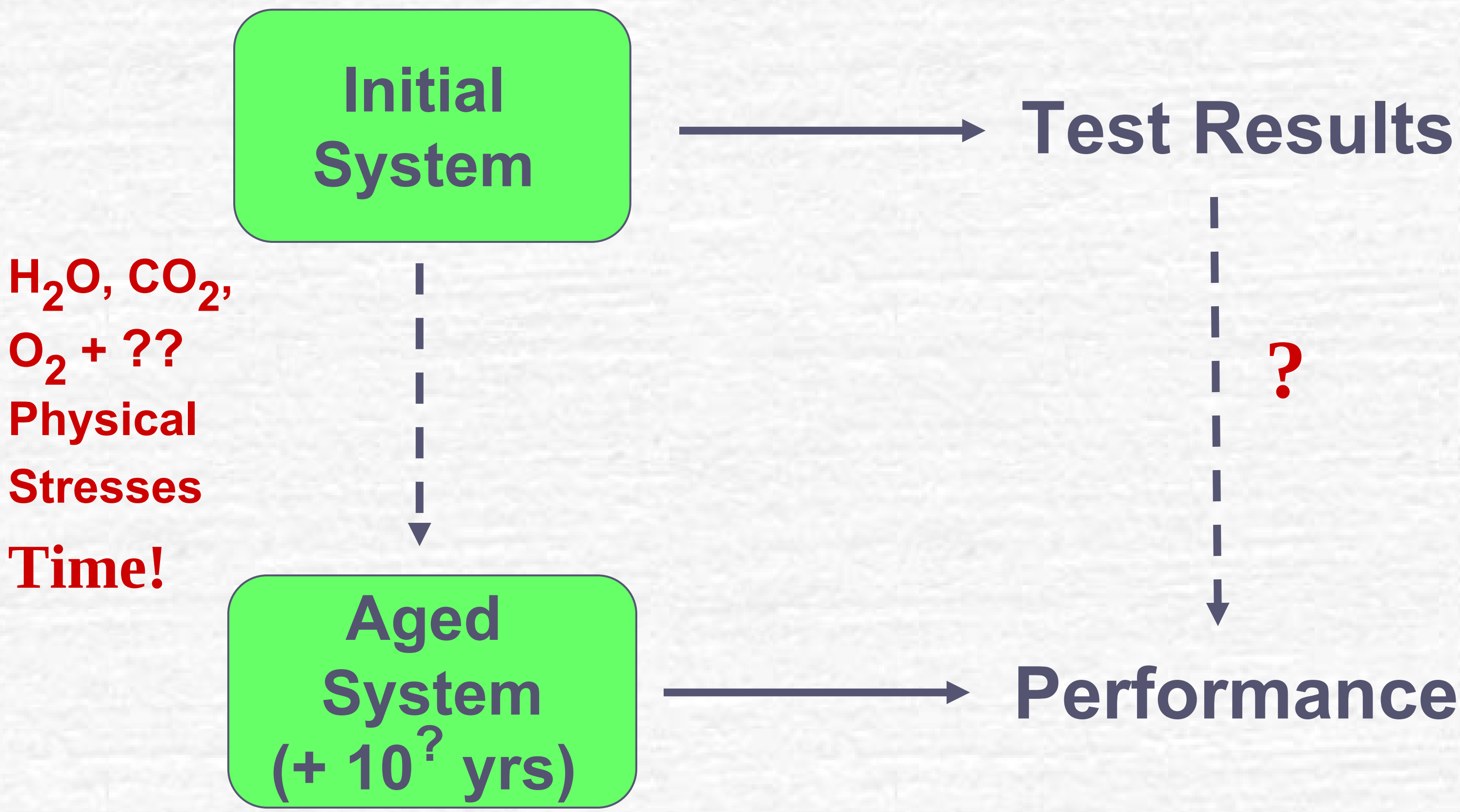
Example - Integrated Use of Testing and Simulation



C_{eq}=f(pH)

$C_{porewater}$= Initial conc.

The Challenge



Sage Advice

- Time scales for experiment and modelling are longer than other steps.
- Steady effort over a longer time is better than crash programs.
- We are in a position to assess data needs and procedures - reliance, it is suggested, should be on modelling for the long term.

F. Glasser, U. of Aberdeen

Sage Advice

- Define objectives realistically.
- Agree implementation.
- Develop and coordinate work packages.
- Conduct actual trials and learn from scale-up.
- Integrate planning with support activities.
- Develop, present and gain approval for plans.

F. Glasser, U. of Aberdeen

Sage Advice

Action check list:

- Matrix properties: evolution of pH and of electroactivity of redox couples.
- Bonding mechanisms of radwaste species in fresh and altered matrices.
- Formulation priorities: establish protocols including changing nature of “cement” and aggregates.
- Scale up effects.
- Cracking and crack healing: impacts on permeation.
- Role of accelerated testing.
- Development of modelling protocols: verification/validation of model predictions and integration of models for “physical” and “chemical” attributes of performance.

F. Glasser, U. of Aberdeen

The Needs

1. Controlling Mechanisms (definition & quantification)

- Geochemical Phases & Aqueous-Solid Partitioning
  - Thermodynamics
- Critical Reactions
  - Kinetics
- Mass transfer relationships
  - Permeability as a function of cracking
  - Aqueous & Gas phase diffusivity as a function of moisture saturation, porosity
  - Formation of reaction fronts, altered/depleted rinds, sharp layers
- Physical deterioration
  - Mechanisms & rates
- Coupling (synergistic & antagonistic) amongst processes

The Needs

2. Standardized & Accepted Test Methods

- Parameter Estimates
  - Leaching (equilibrium, mass transfer)
  - Reducing capacity
  - Permeability for saturated & unsaturated conditions (liquid & gas phases)
  - Structural degradation (Young's modulus, others)
  - Others
- Accelerated Aging
  - Carbonation, oxidation, Sulfate attack, matrix depletion
- Simplified Testing to Support Operations

The Needs

3. Multi-scale Verification of Conceptual Models and Predictions

- Laboratory
- Pilot-scale
- Full-scale (monitoring and evaluation of current systems, e.g., saltstone)

The Needs

4. Guidance & Criteria

- Sample characterization
  - How many samples?
  - What should be measured?
- Waste form formulation & selection
- System design
- Performance Monitoring
- De minimus levels for reuse of D&D materials

The Needs

5. Standardized Performance Models and Databases

- Contaminant Release Model
  - balances mechanistic understanding and practical implementation
- Structural Integrity & Degradation Model
  - balances mechanistic understanding and practical implementation
- Performance Scenarios
  - Tank closure, near surface disposal, D&D
- Parameter and Testing Database
  - Geochemical formalization, thermodynamics, kinetics, mass transfer, other physical & chemical properties
  - Testing data
- Experience Knowledge Base

Good News!

We are not alone!

- Commonality amongst DOE sites and with private sector nuclear facilities
- Commonality with other sectors
  - Waste management
  - Construction
- US & International Experience

There is a vast amount of knowledge, experience and on-going inquiry that should be coordinated for most efficient progress.

Path Forward

- Secure Leadership Commitment
  - Multi-agency collaboration
  - Engagement of research, practitioner and regulatory communities
  - Stakeholder communication
  - Realistic allocation of resources
- Develop Roadmap for Progress
  - Link with key milestones and decisions
  - Link project needs with research, development and standardization progress
- Establish User Groups and Implementation Teams
- Make it happen!

Thank You!

Mark Gilbertson

Deputy Assistant Secretary

Engineering & Technology

Office of Environmental Management